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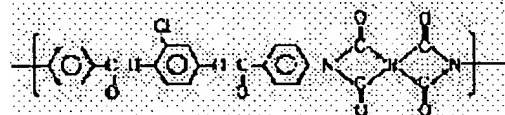
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(54) COPOLYIMIDE AND ITS PRODUCTION

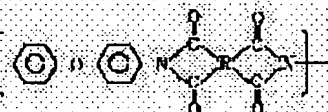
(57)Abstract:

PURPOSE: To provide a copolyimide which has an excellent heat resistance, high mechanical strengths, and low thermal expansion coefficient and water absorption properties and is suitable for a substrate of a fine-patterned flexible printed circuit board.

CONSTITUTION: This copolyimide mainly comprises repeating units represented by formula I and repeating units represented by formula II. In those formulas, R is a tetravalent arom. group.



I



II

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention has a high mechanical strength, a low coefficient of thermal expansion, and low water absorption, and relates to a polyimide copolymer suitable as base materials, such as a fine patternizing flexible-printed-wiring substrate, and its manufacture approach while it is excellent in thermal resistance.

[0002]

[Description of the Prior Art] Conventionally, all the aromatic polyimide that was rich in the flexibility acquired from 4 and 4'-diamino diphenyl ether [which it is known that polyimide resin has the thermal resistance, the chemical resistance, electrical property and mechanical property which was very excellent, and many other outstanding properties, for example, is looked at by JP,36-10999,B], and pyromellitic acid 2 anhydride is known well.

[0003] Since this polyimide resin included the ether linkage which is rich in sturdy nature in a principal chain, though it was all aromatic polyimide, on the other hand, the elastic modulus was low and it had that problem that is rich in flexibility that thermal dimensional stability was bad. Especially the polyimide resin currently used widely conventionally has a coefficient of linear expansion as large as abbreviation $3 \times 10^{-5}/\text{degree C}$, and when thermal dimensional stability is bad and carries out a laminating to a metal etc., since it is easy to produce curvature and curl, and water absorption is remarkable, and is large and dimensional stability and an insulating fall are produced, the problem that electrical characteristics fall has been produced.

[0004] On the other hand, it has the more excellent thermal dimensional stability, and the demand to the polyimide of low hygroscopicity is increasing, maintaining the high thermal resistance which polyimide resin has, and a high mechanical strength in recently, and, for this reason, examination is performed variously. Many measures which aim at the improvement in a mechanical property, thermal dimensional stability, etc. and the improvement to low absorptivity using two or more sorts of aromatic series diamines in this attempt are seen. However, it was not what is satisfied more nearly enough [still] in the thermal dimensional stability and the absorptivity which are a solution technical problem in any [of these measures] case.

[0005] While this invention was made in view of the above-mentioned situation, and has the outstanding thermal resistance and the outstanding mechanical strength, and its thermal dimensional stability is moreover high and having a low coefficient of linear expansion equivalent to a metal, it aims at offering the polyimide copolymer which has low absorptivity, and its manufacture approach.

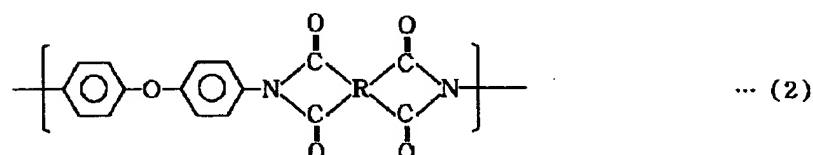
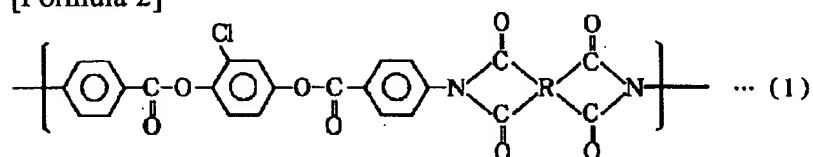
[0006]

[Means for Solving the Problem and its Function] The result of having inquired wholeheartedly in order that this invention person etc. might attain the above-mentioned purpose, Carry out the polymerization of the aromatic series diamine and aromatic series tetracarboxylic dianhydride which use a 1 and 4-screw (4-amino benzoyloxy)-2-chlorobenzene and 4 and 4'-diamino diphenyl ether as a principal component, and a polyamide acid copolymer is obtained. The polyimide copolymer which includes the

repeating unit shown by the following general formula (1) by carrying out the dehydration ring closure of this thermally or chemically, and the repeating unit shown by the following general formula (2) as a main configuration unit, While a polyimide copolymer including the repeating unit the repeating unit preferably shown by the following general formula (1) and whose repeating unit shown by the following general formula (2) are (1) / (2) = 5 / 95 - 90/10 in a mole ratio is obtained It has thermal resistance and a mechanical strength excellent in this polyimide copolymer, and thermal dimensional stability is high and it has a low coefficient of linear expansion equivalent to a metal and low water absorption, a still higher elastic modulus, and flexibility. This sake, The knowledge of being effectively used as base materials, such as a fine patternizing flexible-printed-wiring substrate, is carried out, and it comes to make this invention.

[0007]

[Formula 2]



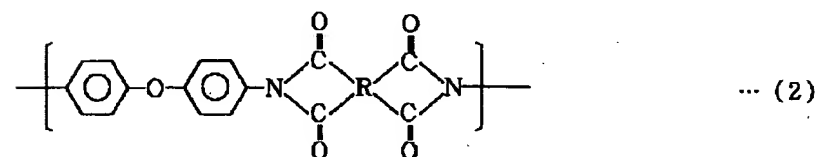
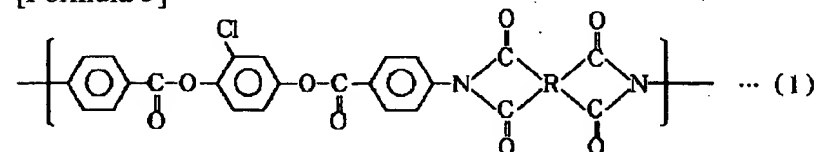
(However, a tetravalent aromatic series radical is shown by the inside R of a formula.)

[0008] Therefore, the polyimide copolymer characterized by this invention including the repeating unit shown by the above-mentioned general formula (1), and the repeating unit shown by the above-mentioned general formula (2) as a main configuration unit, The polyimide copolymer the repeating unit of the above-mentioned formula (1) and whose repeating unit of the above-mentioned formula (2) are (1) / (2) = 5 / 95 - 90/10 in a mole ratio preferably, And carry out the polymerization of the aromatic series diamine and tetracarboxylic dianhydride which use a 1 and 4-screw (4-amino benzoyloxy)-2-chlorobenzene and 4 and 4'-diamino diphenyl ether as a principal component, and a polyamide acid copolymer is obtained. Subsequently, the manufacture approach of the polyimide copolymer which includes the repeating unit shown by the above-mentioned general formula (1) characterized by carrying out a dehydration ring closure for this polyamide acid copolymer thermally or chemically and the repeating unit shown by the above-mentioned general formula (2) as a main configuration unit is offered.

[0009] Hereafter, it explains per this invention and also to a detail. The polyimide copolymer of this invention includes the repeating unit shown by the following general formula (1), and the repeating unit shown by the following general formula (2) as a main configuration unit.

[0010]

[Formula 3]



[0011] The above-mentioned inside R of a formula is a tetravalent aromatic series radical, and this originates in the main frame of the tetracarboxylic dianhydride mentioned later.

[0012] Here, as for the above-mentioned repeating unit (1) and the above-mentioned repeating unit (2), it is desirable in a polyimide copolymer (1) / (2) = 5 / 95 - 90/10, and to exist especially at a rate of 15 / 85 - 80/20 by the mole ratio. If the mole ratio of the above-mentioned repeating unit (1) exceeds 90%, the flexibility of a polyimide copolymer will fall very much. Moreover, if the mole ratio of the above-mentioned repeating unit (2) exceeds 95%, the water absorption of a polyimide copolymer becomes high and the improvement effect of a coefficient of linear expansion and an elastic modulus may not fully be acquired, either.

[0013] moreover, a logarithm [in / when the above-mentioned polyimide copolymer is a polymer of the amount of macromolecules and the viscosity as a polyamide acid copolymer is measured for example, in 0.5g / 100mlDMF / the measurement temperature of 30 degrees C] -- it is desirable that viscosity is 0.5-5.0.

[0014] The polyimide copolymer of this invention can carry out the polymerization of the aromatic series diamine and tetracarboxylic dianhydride which use a 1 and 4-screw (4-amino benzoyloxy)-2-chlorobenzene and 4 and 4'-diamino diphenyl ether as a principal component, can obtain a polyamide acid copolymer, and can manufacture it by next carrying out the dehydration ring closure of this polyamide acid copolymer.

[0015] Here, although it is most desirable as aromatic series diamine to use only 1 and 4-screw (4-amino benzoyloxy)-2-chlorobenzene and 4, and 4'-diamino diphenyl ether, other aromatic series diamine compounds can be used together with such aromatic series diamines. As aromatic series diamine which can be used together, a 4 and 4'-screw (4-amino phenoxy) biphenyl, - diamino diphenyl sulfone, and 4 and 4' 3, 3'-diamino diphenyl sulfone, Screw {4-(4-amino phenoxy) phenyl} sulfone, screw {4-(3-amino phenoxy) phenyl} sulfone, Screw {4-(2-amino phenoxy) phenyl} sulfone, 1, 4-screw (4-amino phenoxy) benzene, 1, 3-screw (4-amino phenoxy) benzene, 1, 3-screw (3-amino phenoxy) benzene, 1, 4-screw (4-aminophenyl) benzene, the screw {4-(4-amino phenoxy) phenyl} ether, 4 and 4'-diamino diphenylmethane, screw (3-methyl-4-aminophenyl) methane, Screw (3-chloro-4-aminophenyl) methane, 3, 3'-dimethoxy -4, 4'-diamino diphenyl, 3, the 3'-dimethyl -4, a 4'-diamino biphenyl, 3, 3'-dichloro -4, a 4'-diamino biphenyl, 2, 2', 5, and 5' - tetra-chloro -4 and 4' - diamino biphenyl -- 3 3'-dicarboxy -4, a 4'-diamino biphenyl, 3, 3'-dihydroxy -4, a 4'-diamino biphenyl, - diamino diphenyl sulfide, and 4 and 4' 3, 3'-diamino diphenyl ether, 3, a - diamino diphenyl ether, and 4' 4, 4'-diamino biphenyl, A 4 and 4'-diamino octafluoro biphenyl, 2, 4-diaminotoluene, A p phenylenediamine, meta-phenylenediamine, 4, and 4'-diamino benzanilide, A - diamino benzanilide, and 4' 4, 3'-diamino benzanilide, [3, and] 2 and 2-screw {4-(4-amino phenoxy) phenyl} propane, 2 and 2-screw {4-(4-amino phenoxy) phenyl} hexafluoropropane, 2 and 2-screw (3-hydroxy-4-aminophenyl) propane, 2, and 2-screw (3-hydroxy-4-aminophenyl) hexafluoropropane, A 9 and 9-screw (4-aminophenyl)-10-hydronalium-anthracene, orthotolidine sulfone, etc. can be mentioned. Furthermore, use of some tetra-amines, such as - biphenyl tetra-amine, 3, 3', and 3, 3', 4, and 4' 4, 4'-tetra-amino diphenyl ether, is also possible. although aromatic series diamines other than a 1 and 4-screw (4-amino benzoyloxy)-2-chlorobenzene are usable among such aromatic series diamines within limits by which the purpose and effectiveness of this invention are attained -- these -- others -- the amount of the aromatic series diamine compound used has desirable use of the amount which does not exceed ten-mol % to all aromatic series diamine compounds, and the amount which does not exceed especially five-mol %.

[0016] As tetracarboxylic dianhydride, concretely Moreover, pyromellitic acid 2 anhydride, - biphenyl tetracarboxylic dianhydride, 2 and 3, 3', 4, and 4'3'4'-biphenyl tetracarboxylic dianhydride, 3, 3', 4, 4'-benzophenone tetracarboxylic dianhydride, 2, 3 and 6, 7-naphthalene tetracarboxylic dianhydride, 1, 4, 5, 8-naphthalene tetracarboxylic dianhydride, 2, and 2-screw (3, 4-dicarboxy phenyl) propane 2 anhydride, Screw (3, 4-dicarboxy phenyl) ethane 2 anhydride, screw (3, 4-dicarboxy phenyl) ether acid 2 anhydride, 1 and 1-screw (3, 4-carboxyphenyl) ethanoic acid 2 anhydride, 3, 4 and 9, 10-perylene tetracarboxylic dianhydride, Benzene - 1, 2, 3, 4-tetracarboxylic dianhydride, 2, 3 and 6, 7-anthracene

tetracarboxylic dianhydride, 1, 2, 7, and 8-phenylene tetracarboxylic dianhydride etc. can be mentioned, and these are independent in a kind, or can use together and use two or more sorts.

[0017] When making above-mentioned aromatic series diamine and aromatic series tetracarboxylic dianhydride react, the polymerization degree of the polyimide copolymer manufactured when it was desirable to mix at a rate of 96-105 mols and it separated from aromatic series tetracarboxylic dianhydride from the above-mentioned range to 100 mols of aromatic series diamine mixture may not go up, and sufficient mechanical strength may be unable to be maintained.

[0018] Moreover, the reaction of aromatic series diamine and aromatic series tetracarboxylic dianhydride can be performed in an organic polar solvent. As an organic polar solvent to be used, for example In this case, sulfoxide system solvents, such as dimethyl sulfoxide, Formamide system solvents, such as N,N-dimethylformamide, N, and N-diethyl formamide, Acetamide system solvents, such as N,N-dimethylacetamide, N, and N-diethyl acetamide, Phenol system solvents, such as pyrrolidone system solvents, such as a N-methyl-2-pyrrolidone, a phenol, o-, m- or p-cresol, a xylenol, a halogenation phenol, and a catechol, Or hexa methyl phospho RUMUAMIDO, gamma-butyrolactone, etc. can be mentioned. These organic polar solvent is independent in a kind, or it is also possible to mix and use two or more sorts, and to use together and use the organic nonpolar solvent of aromatic hydrocarbon systems, such as a xylene and toluene, for the above-mentioned organic polar solvent. Although especially the amount of the above-mentioned organic polar solvent used is not limited, it is desirable to determine the amount of aromatic series diamine, aromatic series tetracarboxylic dianhydride, and the organic polar solvent used that the polyamide acid copolymer obtained by the polymerization reaction is dissolving 10 to 20% especially five to 30% of the weight into an organic polar solvent.

[0019] In addition, an above-mentioned polymerization reaction is 0-70 degrees C, and can obtain a polyamide acid copolymer more effectively for 1 to 50 hours by carrying out at 0-30 degrees C especially for 4 to 24 hours.

[0020] the logarithm at the time of measuring in the measurement temperature of 30 degrees C in 0.5g / 100mlDMF, as mentioned above although especially the viscosity of the polyamide acid copolymer obtained above was not restricted -- it is desirable that viscosity is 0.5 - 5.0 dl/g, and this becomes the index of the molecular weight of a polyimide copolymer.

[0021] The polyamide acid copolymer compounded as mentioned above can manufacture a polyimide copolymer by subsequently performing a dehydration ring closure. Approaches, such as for example, thermal / chemical dehydration ring closure, are employable as the dehydration ring closure approach.

[0022] When carrying out the dehydration ring closure of the polyamide acid copolymer thermally, the approach of heating for 5 - 120 minutes at 200-500 degrees C is suitable.

[0023] Moreover, the approach using a dehydrating agent and a catalyst is suitable to carry out a dehydration ring closure chemically. In this case, as a dehydrating agent, aliphatic series acid-anhydride, aromatic series acid-anhydride, N, and N'-dialkyl carvone imide, a lower-fatty-acid halogenide, a halogenation lower-fatty-acid halogenide, a halogenation lower-fatty-acid anhydride, an allyl compound phosphonic acid dihalogen ghost, a thionyl halogenide, etc. can be mentioned, and these are independent in a kind, or can mix and use two or more sorts. Especially the amount of the above-mentioned dehydrating agent used has desirable 2 - 6 molar quantity about 0.5 to 10 molar quantity per repeat unit of a polyamide acid copolymer.

[0024] In case a catalyst is used, heterocycle type tertiary amines, such as aromatic series tertiary amines, such as aliphatic series tertiary amines, such as triethylamine, and dimethylaniline, a pyridine, beta-picoline, and an isoquinoline, etc. can be mentioned, and these are independent in a kind, or can mix and use two or more sorts. Especially the amount of the above-mentioned catalyst used has desirable 0.1 - 2 molar quantity about 0.01 to 4 molar quantity per repeat unit of a polyamide acid copolymer.

[0025] In order to react the above-mentioned chemical dehydration ring closure, 0.5 - 5 hours is especially desirable at 50-350 degrees C at 20-400 degrees C for 0.2 to 20 hours.

[0026] Moreover, in order to obtain a polyimide copolymer as the shape of a film, the organic polar

solvent of the above-mentioned polyamide acid copolymer is cast or applied to base materials, such as an endless belt, it considers as the shape of film, this film is dried at 100-150 degrees C, and the free-standing film of the polyamide acid copolymer which contains a solvent 10 to 30% is obtained.

Subsequently, after tearing off this film from a base material and fixing an edge, a polyimide film with a thickness of about 10-150 micrometers can be obtained by heating at about 150-250 degrees C, flying a solvent, and forming dehydration imide at 250-500 degrees C further.

[0027] Thus, the polyimide copolymer obtained has a high mechanical strength, a low coefficient of thermal expansion, and low water absorption, and is suitably used as base materials, such as a fine patternizing flexible-printed-wiring substrate, while excelling in thermal resistance.

[0028]

[Example] Although an example and the example of a comparison are shown and this invention is explained concretely hereafter, this invention is not restricted to the following example.

[0029] [Example of reference] The 1 and 4-screw (4-amino benzoyloxy)-2-chlorobenzene was compounded as follows first.

[0030] 1 and 4-dihydroxy-2-chlorobenzene 49.2g (0.340 mols) and triethylamine 81.0g (0.800 mols) were dissolved in tetrahydrofuran 300ml, and the solution which melted tetrahydrofuran 150ml and 139.2g (0.750 mols) of p-nitro benzoyl chlorides in [after cooling at 0 degree C] it was dropped so that the temperature of reaction mixture might become 10 degrees C or less. Then, it returned to the room temperature and stirring was continued for 2 hours.

[0031] Subsequently, after ****(ing) the sludge, washing by the tetrahydrofuran and water and a methanol washing further, it dried and the white crystal of a 1 and 4-screw (4-nitrobenzoyloxy)-2-chlorobenzene was obtained. The yield was 145.9g (96.9% of yield). The rough crystal was recrystallized by N.N-dimethylformamide and the pure article was obtained.

[0032] 1 and 4-screw (4-nitrobenzoyloxy)-2-chlorobenzene 110.7g (0.250 mols) obtained above was inserted in the 1000ml autoclave with Pd/C3g and dimethylformamide 600ml 5%. Stirring was continued until it introduced hydrogen and absorption of hydrogen was no longer accepted, stirring violently at 50 degrees C.

[0033] It ****(ed), after cooling, vacuum concentration of the catalyst was removed and carried out, it flowed into 1000ml of water, and sediment was ****(ed), with water, after washing, reduced pressure drying was carried out and the white solid-state of a 1 and 4-screw (4-amino benzoyloxy)-2-chlorobenzene was obtained. Yield was 91.3g (95.4% of yield). The rough crystal was recrystallized with the mixed solvent of dimethylformamide/methanol, and the pure article was obtained.

[0034] [Example 1] 409.4g of N.N-dimethylformamide was put into the 1000ml flask, and 1, 4-screw (4-amino benzoyloxy)-2-chlorobenzenesg [7.656] (0.020 mols) and 4, and 4'-diamino diphenyl ether 16.019g (0.080 mols) was dissolved for nitrogen gas in N.N-dimethylformamide with the sink. Next, 21.812g (0.100 mols) of pyromellitic acid 2 anhydrides was added, and it was made to react at 25 degrees C for 6 hours.

[0035] next -- since these amic-acid copolymer solutions are thinly extended by the applicator on a glass plate and 110 degrees C dries for 60 minutes among oven -- exfoliating -- an iron frame -- fixing -- 200 degrees C -- subsequently, desolventization imide was formed and 300 degrees C of polyimide films of about 25-micrometer thickness were obtained for 60 minutes for 60 minutes. The property of this polyimide film is shown in Table 1.

[0036] [Example 2] 442.2g of N.N-dimethylformamide was put into the 1000ml flask, and 1, 4-screw (4-amino benzoyloxy)-2-chlorobenzenesg [15.312] (0.040 mols) and 4, and 4'-diamino diphenyl ether 12.014g (0.060 mols) was dissolved for nitrogen gas in N.N-dimethylformamide with the sink. Next, after adding 21.812g (0.100 mols) of pyromellitic acid 2 anhydrides and making it react at 25 degrees C for 6 hours, the polyimide film was obtained by the same approach as an example 1. The property of this polyimide film is shown in Table 1.

[0037] [Example 3] 475.1g of N.N-dimethylformamide was put into the 1000ml flask, and 1, 4-screw (4-amino benzoyloxy)-2-chlorobenzenesg [22.968] (0.060 mols) and 4, and 4'-diamino diphenyl ether 8.010g (0.040 mols) was dissolved for nitrogen gas in N.N-dimethylformamide with the sink. Next, after

adding 21.812g (0.100 mols) of pyromellitic acid 2 anhydrides and making it react at 25 degrees C for 6 hours, the polyimide film was obtained by the same approach as an example 1. The property of this polyimide film is shown in Table 1.

[0038] [Example 4] 508.0g of N.N-dimethylformamide was put into the 1000ml flask, and 1, 4-screw (4-amino benzoyloxy)-2-chlorobenzenesg [30.624] (0.080 mols) and 4, and 4'-diamino diphenyl ether 4.005g (0.020 mols) was dissolved for nitrogen gas in N.N-dimethylformamide with the sink. Next, after adding 21.812g (0.100 mols) of pyromellitic acid 2 anhydrides and making it react at 25 degrees C for 6 hours, the polyimide film was obtained by the same approach as an example 1. The property of this polyimide film is shown in Table 1.

[0039] [Example 1 of a comparison] 540.8g of N.N-dimethylformamide is put into a 1000ml flask, 1 and 4-screw (4-amino benzoyloxy)-2-chlorobenzene 38.280g (0.100 mols) was added with the sink, and nitrogen gas was dissolved in N.N-dimethylformamide. Next, after adding 21.812g (0.100 mols) of pyromellitic acid 2 anhydrides and making it react at 25 degrees C for 6 hours, the polyimide film was obtained by the same approach as an example 1. The property of this polyimide film is shown in Table 1.

[0040] [Example 2 of a comparison] 376.5g of N.N-dimethylformamide is put into a 1000ml flask, 4 and 4'-diamino diphenyl ether 20.024g (0.100 mols) was added with the sink, and nitrogen gas was dissolved in N.N-dimethylformamide. Next, after adding 21.812g (0.100 mols) of pyromellitic acid 2 anhydrides and making it react at 25 degrees C for 3 hours, the polyimide film was obtained by the same approach as an example 1. The property of this polyimide film is shown in Table 1.

[0041] the obtained polyimide film -- a mechanical property, a coefficient of linear expansion, water absorption, and a logarithm -- viscosity was measured. A measuring method is shown below.

Mechanical property ASTM (tensile strength, an elastic modulus, ductility) It measured based on D 882-88.

the product made from Coefficient-of-linear-expansion Vacuum Science and engineering -- the average with a coefficient of linear expansion of 150-200 degrees C was calculated with the programming rate 5 (a part for degree-C/) using thermal-analysis meter TMA-7000.

The water absorption polyimide film was left in RH 90% for 24 hours, the weight before and behind that was measured, and water absorption was searched for.

a logarithm -- it is the viscosity polyamide acid copolymer concentration of 0.5g / 100mlDMF, and computed in the following formula from the result measured on the conditions whose measurement temperature is 30 degrees C.

a logarithm -- viscosity = -- the polymer concentration in a natural logarithm (viscosity of the viscosity/solvent of a solution)/solution -- the measurement result of these is shown in Table 1.

[0042]

[Table 1]

	実 施 例				比 較 例	
	1	2	3	4	1	2
ピロメリット酸 二無水物 (g)	21.812	21.812	21.812	21.812	21.812	21.812
(I) 1,4-ビス(4-ア ミノベンゾイルオ キシ)-2-クロ ロベンゼン (g)	7.656	15.312	22.968	30.624	38.280	0
(II) 4,4'-ジアミノジ フェニルエーテル (g)	16.019	12.014	8.010	4.005	0	20.024
(I) / (II) (モル比)	20/80	40/60	60/40	80/20	100/0	0/100
引 張 強 度 (kgf/mm ²)	18.5	18.3	18.0	17.0	非 常 に 脆 く 製 膜 不 可	18.4
引 張 伸 度 (%)	41	25	18	12		62
引 張 弾 性 率 (kgf/mm ²)	347	398	475	544		275
線 膨 張 係 数 ($\times 10^{-6}$ cm/cm/°C)	1.72	1.40	0.99	0.58		2.80
吸 水 率 (%)	1.7	1.6	1.5	1.5		2.2
対 数 粘 度 (dl/g)	1.6	1.3	1.4	1.7	1.5	1.4

[0043]

[Effect of the Invention] When excelled in many properties, such as thermal resistance, since the mechanical strength has the same low coefficient of linear expansion as a metal, and low water absorption highly, the polyimide film constituted with the polyimide copolymer of this invention is suitably used as film materials, such as an electrical insulation material and a fine patternizing flexible-printed-wiring substrate.

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